IN THE U.S. PATENT AND TRADEMARK OFFICE

Applicants : Ohashi et al.

Serial No. : 10/541,776

Filed : July 8, 2005

For : SILICA-CONTAINING LAMINATED STRUCTURE, AND COATING COMPOSITION FOR USE IN FORMING A POROUS SILICA LAYER

Art Unit : 1794

Examiner : Victor S Chang

DECLARATION UNDER 37 C.F.R. 1.132

I, Takaaki Ioka, a Japanese citizen residing at 381-12, Miyajima, Fuji-shi, Shizuoka-ken, Japan declare and say:

I took a master course majoring in chemistry at Graduate School of Science, Tohoku University, Japan, and I was graduated therefrom in March 1996.

I entered Asahi Kasei Kabushiki Kaisha in April 1996. I was engaged in the research and development of electronic industry materials from 1996 to June 2002. Then, I was engaged in the research and development of optical thin film to September 2005. Then, I have been engaged in the research and development of electronic industrial materials to date.

I am one of the applicants of the above-identified application and I am well familiar with the present case.

I have read and understood the Office Action dated June 4, 2009 and the reference cited therein.

I carried out Examples 1 to 21 and Comparative Examples 1 to 7 of the present application, and the results are as

described on pages 74 to 112 of the specification of the present application.

I have made observations on the difference in porous structure between the porous silica layer of the laminated structure of the present invention and the porous silica layer of the laminated structure according to **Takahashi** (US 6251523), and the influence of the difference in porous structure on the optical properties of the laminated structures, referring to Example 21 and Comparative Example 6 of the present application. The observations are described in a paper attached hereto and marked "Exhibit 1".

From the observations of Exhibit 1, it can be fairly concluded:

(1) that, it is apparent that, in Example 21 (present invention) where the porous silica layer is formed by drying the coating composition containing moniliform silica strings at 120 °C for 2 minutes, the resultant laminated structure satisfies the specific large pore characteristics defined in claim 1 of the present application, and exhibits an advantageously low reflectance (0.05 %) without scarifying the strength of the porous silica layer;

(2) that, on the other hand, in Comparative Example 6 of the present application (corresponding to **Takahashi**) where the porous silica layer is formed by heating the same coating composition as used in Example 21 at 500 °C for 1 hour, the resultant laminated structure does not satisfy the specific large pore characteristics as defined in claim 1 of the present application, and exhibits a high reflectance (0.45 % which is almost 10 times higher than that in Example 21); and

(3) that, from items (1) and (2) above, it is apparent that the specific pore characteristics as defined in claim 1 of the present application are essential for achieving a very low reflectance without scarifying the strength of the porous silica layer, and that such specific large pore characteristics cannot be achieved when the coating composition containing moniliform silica strings is heated at a very high temperature (such as 500 °C) as in the Examples of **Takahashi**.

The undersigned petitioner declares that all statements made herein of his own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

Date: December 2, 2009

Takaaki Ioka

Takaaki Ioka

Exhibit 1

Observations on the difference in porous structure between the porous silica layer of the laminated structure of the present invention and the porous silica layer of the laminated structure according to **Takahashi** (US 6251523), and the influence of the difference in porous structure on the optical properties of the laminated structures, referring to Example 21 and Comparative Example 6 of the present application

---

1. OBJECT OF THE OBSERVATIONS

In the Office Action dated June 4, 2009, the Examiner rejects the claims of the present application as being unpatentable over **Lange** (US 4816333) in view of **Takahashi** (US 6251523). Specifically, the Examiner takes a position that **Takahashi** teaches the use of moniliform silica strings for forming a porous silica layer exhibiting excellent optical performance on a glass substrate, and that **Takahashi** also teaches that such a porous silica layer may be formed by drying at a temperature in the range of room temperature to 200 °C for 1 minute to 2 hours and, hence, is combinable with **Lange** which describes the use of a polymer substrate, so that the present invention is obvious over the combination of **Lange** and **Takahashi**.

However, in the present invention, the presence of large pores (P) (having a pore opening area larger than the average value of the respective maximum cross-sectional areas of the primary silica particles) and the specific total value of large pore opening areas (a_1) (at least 3σ larger than average

value (a_2)) represented by formula (1) in claim 1 are essential for simultaneously achieving excellent optical properties and strength. (These features are hereinafter, collectively referred to as "specific large pore characteristics".) On the other hand, **Takahashi** has **no** teaching or suggestion about such specific large pore characteristics and, let alone, the excellent effects achieved thereby. In fact, in the Examples of **Takahashi**, the silica coating formed does not satisfy such specific large pore characteristics as defined in claim 1 of the present application.

For substantiating the above argument, observations are made below, referring to Example 21 and Comparative Example 6 of the present application.

2. OBSERVATIONS BASED ON EXAMPLE 21 AND COMPARATIVE EXAMPLE 6 OF THE PRESENT APPLICATION

2-1. Requirements in claim 1 of the present application

The above-mentioned specific large pore characteristics defined in claim 1 of the present application are as follows:

> " wherein the pores of said at least one porous silica layer include pores (P), each of said pores (P) having a pore opening area which is larger than the average value of the respective maximum cross-sectional areas of said primary silica particles, wherein said pore opening areas of said pores (P) are measured with respect to the pore openings in the surface or cross-section of said porous silica layer,

wherein a part or all of said pores (P) have their respective pore opening areas (a_1), each of said pore opening areas (a_1) being independently at least 3σ larger than the average value (a_2) of the respective maximum cross-sectional areas of said primary silica particles, wherein said pore opening areas (a_1) are measured with respect to the pore openings in the surface or cross-section of said porous silica layer, and wherein σ represents the standard deviation of the measured values of the maximum cross-sectional areas of said primary silica particles, and

wherein the total ($S_{(a2+3\sigma)}$) of said pore opening areas (a_1) of said pores (P) and the total (S) of pore opening areas of all pores of said porous silica layer as measured with respect to the pore openings in the surface or cross-section of said porous silica layer satisfy the following formula (1):

$$(S_{(a2+3\sigma)})/(S) \geq 0.5 \qquad (1).''$$

2-1. Example 21 and Comparative Example 6 of the present application

In Example 21 (page 99, line 18 to page 103, line 1 of the present specification) and Comparative Example 6 (page 103, line 3 to page 104, line 15 of the present specification), a porous silica layer was formed on a substrate using the same coating composition containing moniliform silica strings. However, Example 21 and Comparative Example 6 are different in the following points.

In Example 21, a PET film was used as a substrate, and the coating composition on the substrate was subjected to "drying at 120 °C for 2 minutes using a forced convection

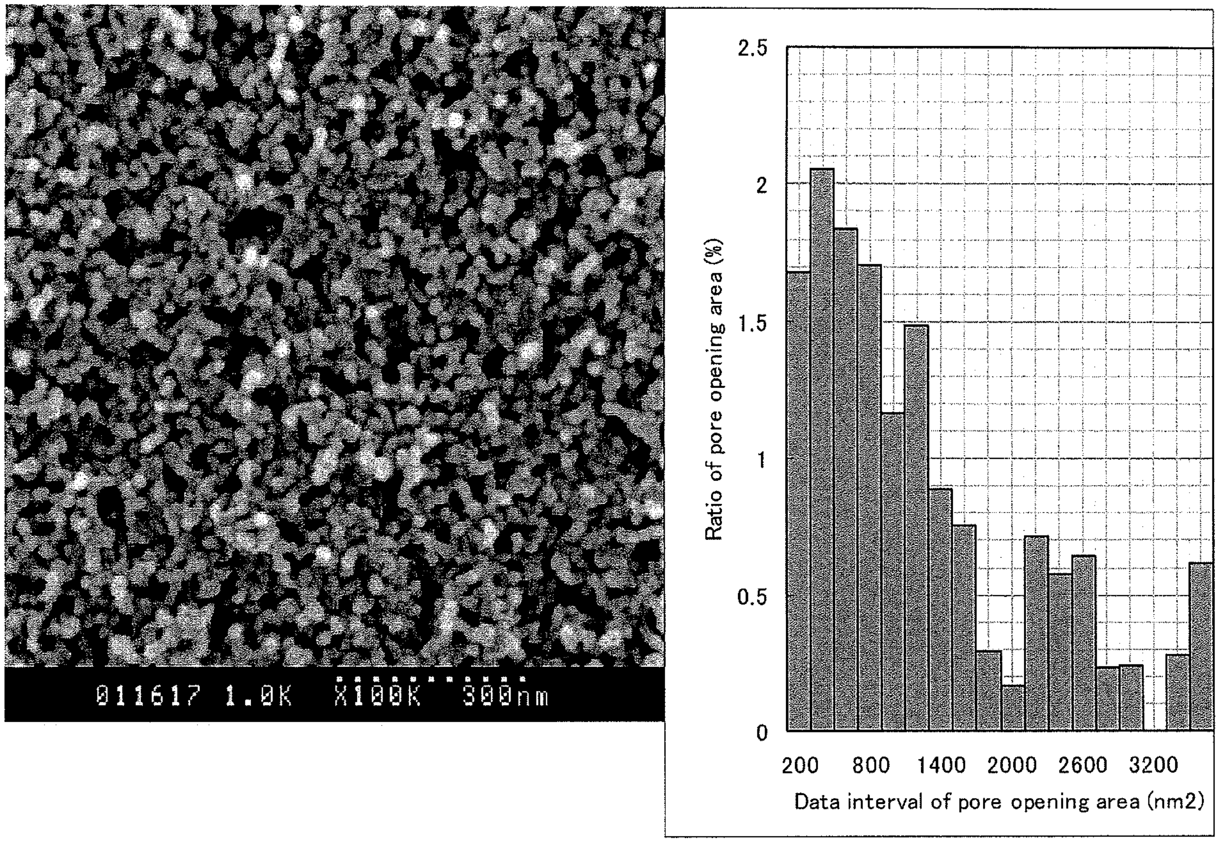
oven" (page 100, lines 18 to 23 of the present specification).

On the other hand, in Comparative Example 6, a glass substrate was used, and the coating composition on the glass substrate was "dried at 120 °C for 2 minutes using a forced convection oven, followed by heating using a muffle furnace at 250 °C for 30 minutes, and then at 500 °C for 1 hour" (page 103, lines 6 to 11 of the present specification).

2-2. Results of Example 21 and Comparative Example 6, and observations thereon

A photomicrograph of the surface of the porous silica layer formed in Example 21 and a graph showing the distribution of pore opening areas obtained from an image analysis of the photomicrograph are shown in Fig. 5' below (the photomicrograph and the graph are obtained by the methods described at page 101 of the present specification).

Fig. 5′ (Example 21)

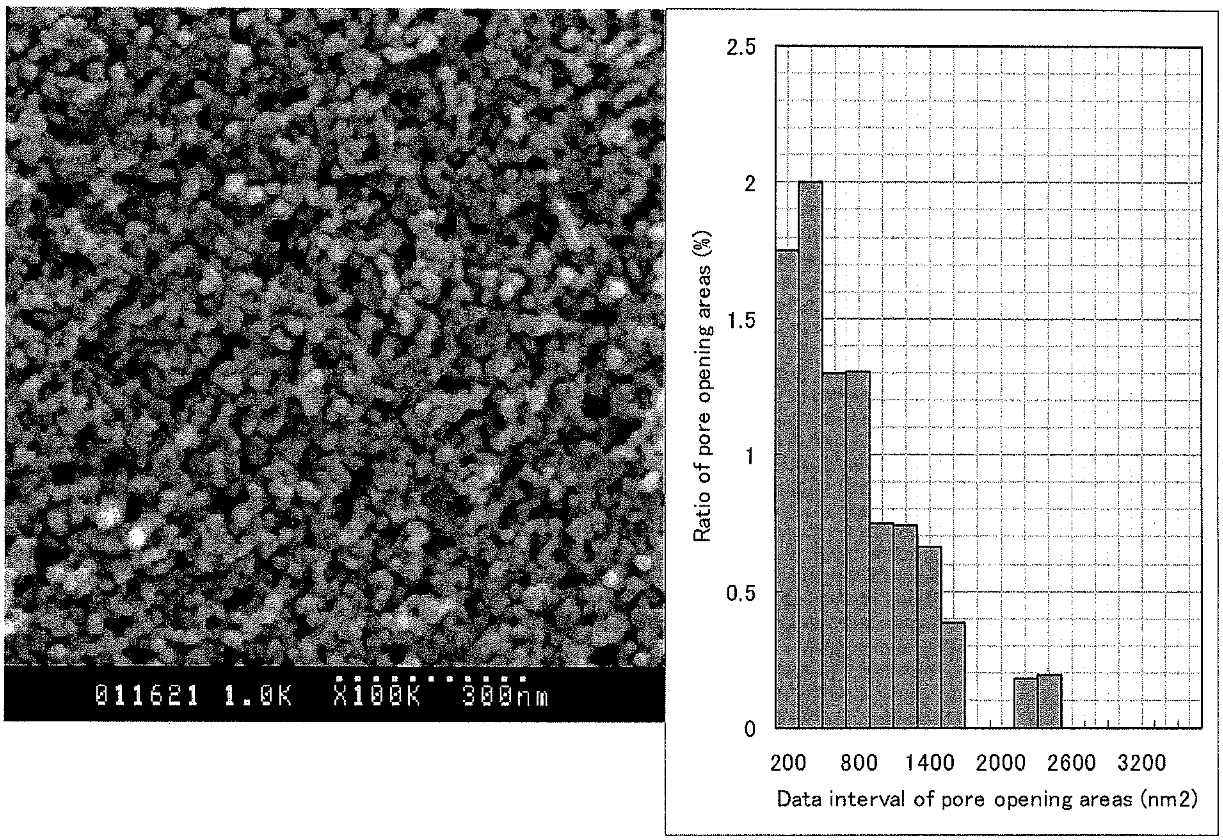


As described at page 102, line 21 to page 103, line 1 of the present specification, in Example 21, it was found that the total ratio (S) of pore opening areas of all pores in the photomicrograph was 20.08 %, that the total ratio ($S_{(a2+3\sigma)}$) of pore opening areas of pores each having a pore opening area which is ($a_2 + 3\sigma$) or more was 13.73 %, and that ($S_{(a2+3\sigma)}$)/(S) was 0.68. Thus, the porous silica layer formed in Example 21 has the specific large pore characteristics defined in claim 1 of the present application.

On the other hand, a photomicrograph of the surface of

the porous silica layer formed in Comparative Example 6 and a graph showing the distribution of pore opening areas obtained from an image analysis of the photomicrograph are shown in Fig. 7′ below (the photomicrograph and the graph are obtained by the methods described at page 101 of the present specification).

Fig. 7′ (Comparative Example 6)



As described at page 104, lines 9 to 15 of the present specification, in Comparative Example 6, it was found that the total ratio (S) of pore opening areas of all pores in the photomicrograph was <u>11.93 %</u>, that the total ratio ($S_{(a2+3\sigma)}$) of

pore opening areas of pores each having a pore opening area which is ($a_2 + 3\sigma$) or more was 4.87 %, and that $(S_{(a2+3\sigma)})/(S)$ was 0.41. Thus, the pore silica layer formed in Comparative Example 6 does not satisfy the specific large pore characteristics defined in claim 1 of the present application.

In connection with the above, it should be noted that, in all of the Examples of **Takahashi**, the coating composition on a glass substrate is heated at a temperature of 500 °C or higher, namely "500 °C for 1 hour" (col. 8, lines 47 to 52 etc.) except for the "Second Embodiment" in which heating was done "for 15 minutes at 570 °C" (col. 11, lines 20 to 25). As long as such high temperature heating is performed, the formation of specific large pores in specific amounts as recited in claim 1 of the present application is impossible due to the heat shrinkage of the silica matrix of the porous silica layer, and the pore structure of the resultant porous silica layer should be substantially the same as in Comparative Example 6 of the present application.

Further, attention is drawn to the following point. As apparent from Fig. 7′ above, the porous silica layer obtained in Comparative Example 6 has "a large number of gaps of 5 to 20 nm width" as described at col.2, lines 20 to 23 of **Takahashi**; nevertheless, the specific large pore characteristics are not satisfied. More specifically, a gap of 20 nm is considered to correspond to a pore opening having an area of slightly larger than 314 nm^2 (which is calculated on the assumption that the gap of 20 nm is a spherical pore having a

diameter of 20 nm). From Fig. 7' above, it is apparent that the porous silica layer formed in Comparative Example 6 has a large number of pores having an area of about 314 nm^2 and, hence, the porous silica layer formed in Comparative Example 6 has "a large number of gaps of 5 to 20 nm width".

From the above, it is crystal-clear that the specific large pore characteristics defined in claim 1 of the present application is not taught or suggested in **Takahashi**.

The optical properties in Example 21 and Comparative Example 6 are, respectively, described at page 100, lines 23 to 25 and page 103, lines 11 to 14 of the specification of the present application, which are shown in Table A below together with pore requirement in claim 1 of the present application, and the pore characteristics in Example 21 and Comparative Example 6.

Table A

	Total ratio (S) of pore opening areas of all pores	Total ratio ($S_{(a2+3\sigma)}$) of pore opening areas of pores each having a pore opening area which is (a_2 + 3σ) or more	($S_{(a2+3\sigma)}$)/(S)	Minimum reflectance	Haze
Claim 1	-	-	$\geq$ 0.5		
Ex. 21	20.08 %	13.73 %	0.68	0.05 %	0.5 %
Comp. Ex. 6	11.93 %	4.87 %	0.41	0.45 %	0.4 %

Thus, the reflectance in Example 21 is almost 1/10 of the reflectance in Comparative Example 6 (corresponding to **Takahashi**).

In this connection, it should be noted that, in the Exam-

ples of **Takahashi**, the reflectance is 0.7 % at the lowest (see Table 9 on col. 6) which is higher than but is relatively close to the reflectance (0.45 %) in Comparative Example 6 of the present application.

In addition, it should be noted that the pencil hardness was not measured in Example 21 and Comparative Example 6 for the following reason. Example 21 and Comparative Example 6 were designed so as to compare the porous structures of the porous silica layer according to the present invention and that of the porous silica layer according to the prior art, such as **Takahashi**; therefore, each of Example 21 and Comparative Example 6 was conducted without using a hard coat layer and on a larger scale than the other Examples. However, since Example 21 is done using the same raw materials as in the other Examples in ratios overlapping those in the other Examples under conditions (such as temperature and time for drying the coating composition) overlapping with those in the other Examples, it is apparent that, when the laminated structure obtained in Example 21 of the present application is produced using a hard coat layer on a similar scale to those in the other Examples, the laminated structure has a pencil hardness at the same level as those (H or 2H) in the other Examples of the present application.

3. CONCLUSION

From the above observations, it is apparent that, in Example 21 (present invention) where the porous silica layer is

formed by drying the coating composition containing moniliform silica strings at 120 °C for 2 minutes, the resultant laminated structure satisfies the specific large pore characteristics defined in claim 1 of the present application, and exhibits an advantageously low reflectance (0.05 %) without scarifying the strength of the porous silica layer.

On the other hand, in Comparative Example 6 of the present application (corresponding to **Takahashi**) where the porous silica layer is formed by heating the same coating composition as used in Example 21 at 500 °C for 1 hour, the resultant laminated structure does not satisfy the specific large pore characteristics as defined in claim 1 of the present application, and exhibits a high reflectance (0.45 % which is almost 10 times higher than that in Example 21).

From the above, it is apparent that the specific pore characteristics as defined in claim 1 of the present application are essential for achieving a very low reflectance without scarifying the strength of the porous silica layer, and that such specific large pore characteristics cannot be achieved when the coating composition containing moniliform silica strings is heated at a very high temperature (such as 500 °C) as in the Examples of **Takahashi**.